

VI.11 Modeling and Design for a Direct Carbon Fuel Cell with Entrained Fuel and Oxidizer

Alan A. Kornhauser (Primary Contact), Ritesh Agarwal
Virginia Tech, Mechanical Engineering
Mail Code 0238
Blacksburg, VA 24061-0238
Phone: (540) 231-7064; Fax: (540) 231-9100; E-mail: alkorn@vt.edu

DOE Project Manager: Travis Shultz
Phone: (304) 285-1370; E-mail: Travis.Shultz@netl.doe.gov

Objectives

- Develop a fuel cell concept in which the anode and cathode are electrically connected porous beds through which electrolyte flows with fuel and oxidizer entrained.
- Develop preliminary solutions to design problems such as electrode construction, gas-solid-electrolyte separation, and balance-of-plant design.
- Model overall plant mass and energy balances.
- Model processes within the electrodes.
- Develop to a stage where potential can be evaluated and research needs determined.

Approach

- Develop cell design.
- Develop balance-of-plant design.
- Model mass and energy balances for the plant.
- Model mass transfer, charge transfer, and chemical kinetics within the cell.
- Determine performance of cell and plant designs. Compare with alternate systems.

Accomplishments

- Selected correlations, from those available in the literature, for mass transfer, reaction rates, and electronic and ionic electrical resistances.
- Modeled performance of cell electrodes with a set of simultaneous differential equations.
- Developed Matlab computer code to solve differential equation set.
- Used electrode models to determine performance of cell design.
- Used electrode models for first-cut optimization of cell design.
- Compared performance with that of alternate direct carbon fuel cell designs.
- Developed design for prototype direct carbon fuel cell.

Future Directions

- Modify design to improve performance.
- Build and test prototype direct carbon fuel cell.

Introduction

Modern fuel cell development has concentrated on compact fuel cell designs in which immobile electrolyte is contained between porous membrane electrodes. Fuel and oxidizer are supplied to the electrodes on opposite sides of the electrolyte, and all reactions take place on the electrolyte-wetted surfaces of the electrode membrane. This design is effective for a compact cell using gaseous fuel, but it has serious limitations for utility-scale power generation using coal.

An alternate concept is proposed. In this concept, the anode and cathode are electrically connected porous beds. Molten salt electrolyte, with carbon fuel and oxidizer entrained, is pumped through the porous beds.

This fuel cell design can use impure solid fuel (coal or coke) and offers economies of scale for utility-size plants. It is particularly attractive since no microporous sintered membranes (both expensive and subject to fouling) are used.

Approach

The technology is more like a chemical refinery or an electrochemical plant than like a membrane electrode fuel cell. Accordingly, design techniques are those used for chemical reactors and electrochemical plants.

Initial designs for the cells and for the balance of plant have been developed. Techniques from the chemical engineering literature have been used to model these designs. The models have been used to predict the performance of the designs, to optimize them, and to design a laboratory prototype cell.

The reactions in the direct carbon molten carbonate fuel cell, the proposed cell design, and the proposed balance of plant design have been described previously [1][2].

One-dimensional models of the anode and cathode beds were developed, as shown schematically in Figure 1. For the anode, there were five dependent variables: CO_2 flux in the gas phase, CO_2 flux in the liquid phase, C flux as particles entrained in the liquid, ionic (CO_3^-) current in the

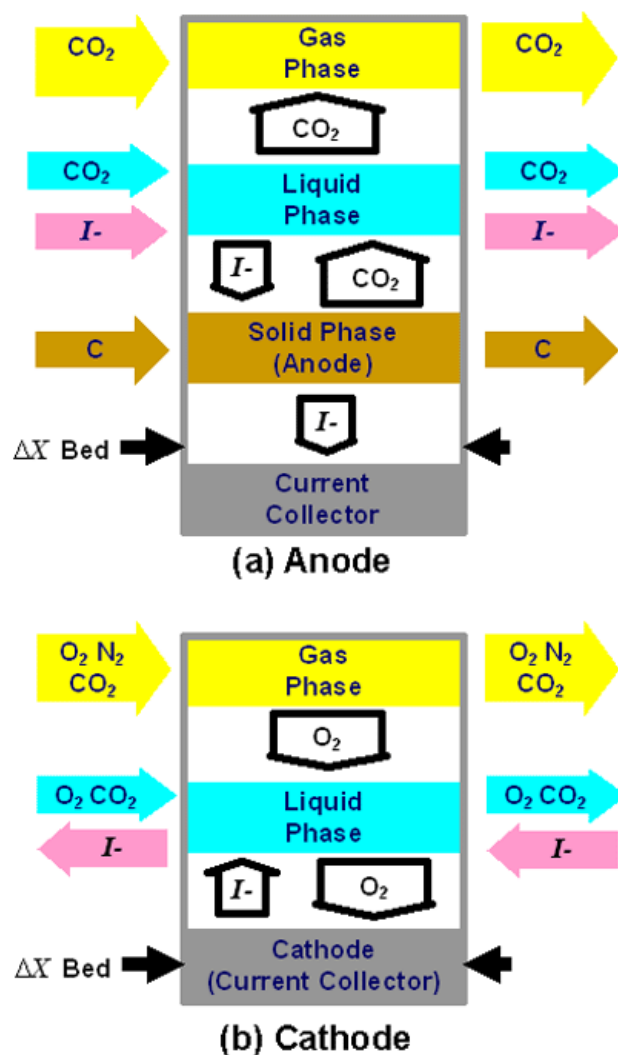


Figure 1. Schematic of Anode and Cathode Models Showing Independent and Dependent Variables

liquid phase, and electric potential. For the cathode, there were six dependent variables: O_2 and CO_2 fluxes in the gas phase, O_2 and CO_2 fluxes in the liquid phase, ionic (CO_3^-) current in the liquid phase, and electric potential. Depth in the bed, x , was the independent variable for both electrodes. Differential equations (five for the anode and six for the cathode) were written for the changes in dependent variables, with terms in the equations based on liquid-gas and liquid-solid mass transfer, liquid ionic conductivity, solid electronic conductivity, and electrode surface reaction rates.

The simultaneous differential equations were written in *Matlab* [3] and solved using the *Matlab*

ordinary differential equation (ODE) solvers. Initial conditions were mole fluxes, current, and voltage at the electrode face. If satisfactory initial conditions were selected, the equations would be stepped forward until the ions representing the initial condition current had been created (cathode) or destroyed (anode). The electrode depth when the current flux went to zero was the minimum required depth for that operating condition. Unsatisfactory initial conditions – due to inadequate initial voltage or reactant flux – would result in overpotential decreasing to zero or reactants being exhausted before current flux went to zero.

Results

The model was used to predict the performance of the cell over a range of operating conditions.

Figure 2 shows voltage profiles for the anode and cathode at a typical operating condition: total

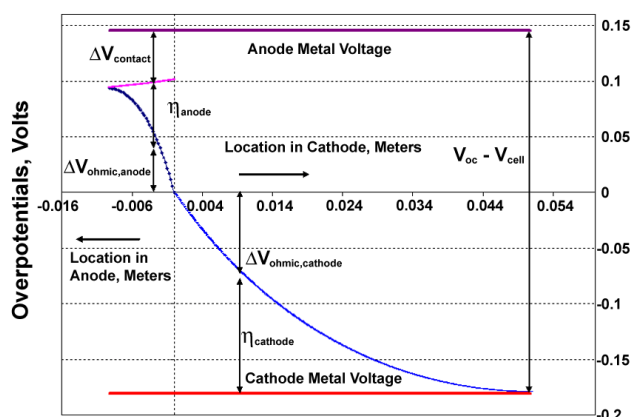


Figure 2. Variation in Overpotentials with Cathode and Anode Bed Depth

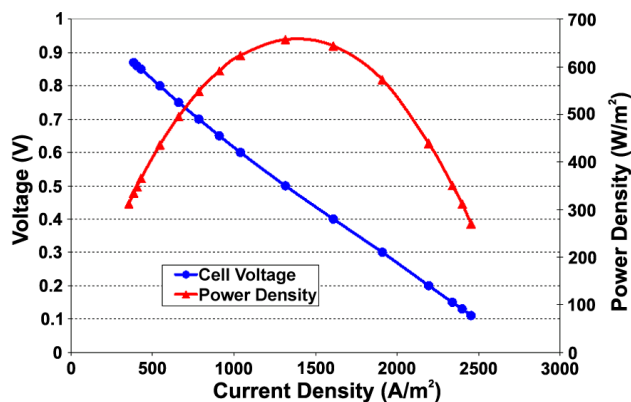


Figure 3. Variation in Output Voltage and Power Density with Bed Face Current Density

potential losses of 0.326 volts, with 0.180 volts assigned to losses in the cathode and 0.146 volts assigned to losses in the anode. At the cathode inlet, losses are all from overcoming mass transfer and chemical overpotentials. At the anode inlet, electron contact resistance between the carbon particles and the current collector is added to these losses. For both electrodes, ionic resistance losses become dominant as distance into the electrode increases, until (at 0.0508 m for the cathode and 0.0092 m for the anode) there is no chemical or diffusion overpotential remaining to drive reactions.

The model was run for various total voltage losses to determine both the maximum ionic current flux and the optimum division of losses between anode and cathode for each case. The results of this (for a single packing type, temperature, and reactant flux) are shown in Figure 3. The cell is seen to attain a maximum power density of 658 W/m^2 at a current density of 1315 A/m^2 and an output potential of 0.5 volts. Since the open circuit potential of the direct carbon molten carbonate cell is 1.026 volts, this corresponds to an efficiency of 49%. Higher efficiencies are obtained at lower power densities.

Design parameters were varied to determine their effect on cell performance. The most important parameter was cell temperature. Figure 4 shows the effect of cell temperature on output power density with other parameters held constant. Output increases dramatically with increasing temperature. This high output at high temperature requires that kinetics favor CO_2 instead of CO , as observed by Weaver et al [4] and Cherepy et al [5].

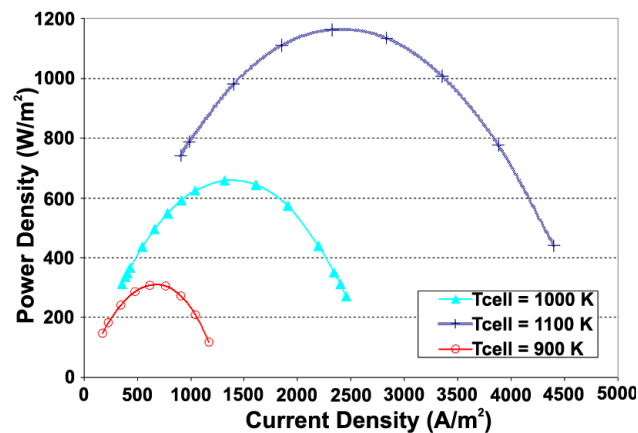


Figure 4. Variation in Output Power Density with Cell Temperature

Conclusions

The analysis performed has both encouraging and discouraging results.

The encouraging result is that the design appears to function basically as expected, allowing construction of a direct carbon fuel cell without microporous electrodes or separating membranes.

The discouraging result is that resistance losses limit the effective depth of the porous bed electrodes to a few centimeters. This, in turn, limits the current density. Predicted current densities are considerably smaller than those demonstrated at Lawrence Livermore National Laboratory on a direct carbon cell using a microporous membrane cathode [5].

The original design concept was based on considerable porous bed depth with relatively small bed face area. Lower bed depths and lower current densities require a modified design with shallow beds with larger face area. Various concepts for this modified design are being developed.

FY 2005 Publications/Presentations

1. Alan A. Kornhauser and Ritesh Agarwal, "Modeling and Design for a Direct Carbon Fuel Cell with Entrained Fuel and Oxidizer – Final Report," Virginia Tech / U.S. DOE, 2005.

References

1. R. Agarwal and A.A. Kornhauser, "Energy Balance for a Direct Carbon Molten Carbonate Fuel Cell," Proceedings of the 2004 ASME Heat Transfer / Fluids Engineering Summer Conference, Charlotte, NC, July 11-15, 2004, Paper HT-FED2004-56887.
2. A.A. Kornhauser and R. Agarwal, "Modeling and Design for a Direct Carbon Fuel Cell with Entrained Fuel and Oxidizer – FY 2004 Annual Progress Report," Virginia Tech / U.S. DOE, 2004.
3. *Matlab Release 14* software package, The Math Works, Woburn, MA, 2004.
4. R.D. Weaver, L. Tietz, and D. Cubicciotti. Direct Use of Coal in a Fuel Cell: Feasibility Investigation. Technical Report EPA-650/2-75-040, SRI International, Menlo Park, California, 1975.
5. N.J. Cherepy, R. Krueger, K.J. Fiet, A.F. Jankowski, and J.F. Cooper, "Direct Conversion of Carbon Fuels in a Molten Carbonate Electrolyte," *Journal of the Electrochemical Society*, 2004.

